Supplementary Data (SD)

SD 1. Fluid Film Lubrication Regime

To identify the regime to which a specific tribological contact belongs, two dimensionless viscosity and elasticity parameters must be evaluated by the parameters g_V and g_E:

Dimensionless viscosity parameter $$g_V = \frac{GW^3}{U^2} \quad (1)$$

Dimensionless elasticity parameter $$g_E = \frac{W^{\frac{8}{3}}}{U^2} \quad (2)$$

where U, G and W are the respective dimensionless parameters for speed, material, and load:

Dimensionless speed parameter $$U = \frac{\eta_0 u}{E'R_x} \quad (3)$$

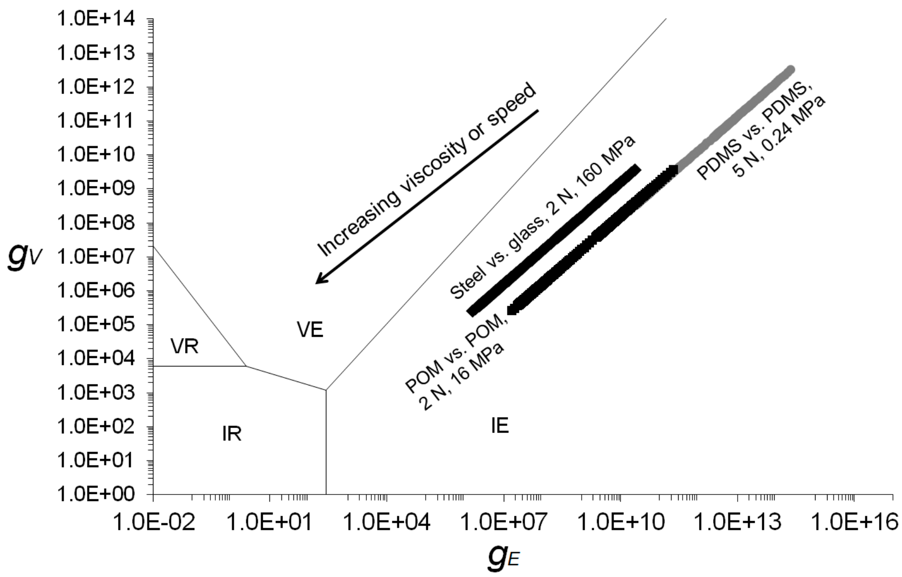
Dimensionless material parameter $$G = \xi E' \quad (4)$$

Dimensionless load parameter $$W = \frac{w}{E'R_x^2} \quad (5)$$

Ellipticity parameter $$k = \frac{2}{\pi}\left(\frac{R_y}{R_x}\right)^{2\pi} \quad (6)$$

In these equations, η_0 is the lubricant's viscosity at atmospheric pressure, u is the mean speed, E' is the reduced Young's modulus, w is the applied load, R_x is the radius in the x direction, R_y is the radius in the y direction, and ξ designates the pressure viscosity coefficient. For circular contacts, the ellipticity parameter (k) is approximated to 1. The Figure S1 below shows that all three different types of contacts in this study belong to isoviscous-elastic (IE) regime for all speeds at 20 °C:

Figure S1. Fluid film lubrication regime, for the three types of contacts, Steel *vs.* glass (*h*-**HL**), POM *vs.* POM (*h*-**HB**) and PDMS *vs.* PDMS (*s*-**HB**/*s*-**HL**). The shown plots are for all speeds (10–1200 mm/s) at 20 °C. However, all the three types of contacts lie in the isoviscous-elastic (IE) regime in the plot even temperature is varied from 1 °C to 90 °C (not shown).



SD 2. Temperature Dependence of Pdms's Young's Modulus

For elastomers, the influence of temperature on the Young's modulus (E) can be modeled by the following Equation [1]:

$$E = 3 \cdot G = 3 \cdot n_c \cdot k_B \cdot T \quad (7)$$

where G, n_c, k_B and T is shear modulus, number of crosslinks per unit volume of an elastomer, Boltzmann's constant, and absolute temperature. In cured PDMS from Sylgard 184® at 10:1 base: crosslinker wt. ratio, the number of crosslinks is $1.6 \times 10^{26}/m^3$ [2]. In Figure SD2-A, E *vs.* temperature is plotted.

At the lowest temperature, 1 °C, E = 1.82 MPa, and the highest temperature, 90 °C, E = 2.41 MPa. The resulting effective Young's modulus of the 2 mm PDMS on the 5 mm steel disc substrate is then 6.37 and 8.43 MPa, at 1 and 90 °C, respectively. The difference in film thickness due to the change in Young's modulus as a result of temperature change is $E \propto (E_2/E_1)^{-0.45} = (8.43\ \text{MPa}/6.37\ \text{MPa})^{-0.45} = 0.88$, thus only a 12% difference in film thickness. The contact pressure change due to the difference in Young's modulus at difference temperature goes from 0.235 MPa to 0.245 MPa. Figure SD2-B below shows the calculated Stribeck parameter (λ) *vs.* mean speed at the different temperatures, compensated and non-compensated for the changes in Young's modulus. The difference is ignorable.

Figure S2-A. *E vs.* temperature from 1 to 100 °C.

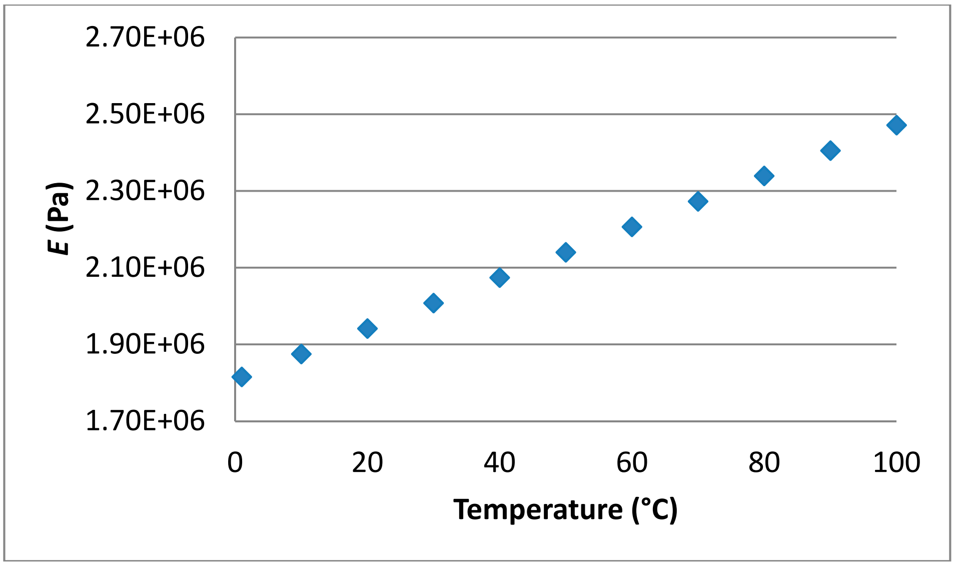
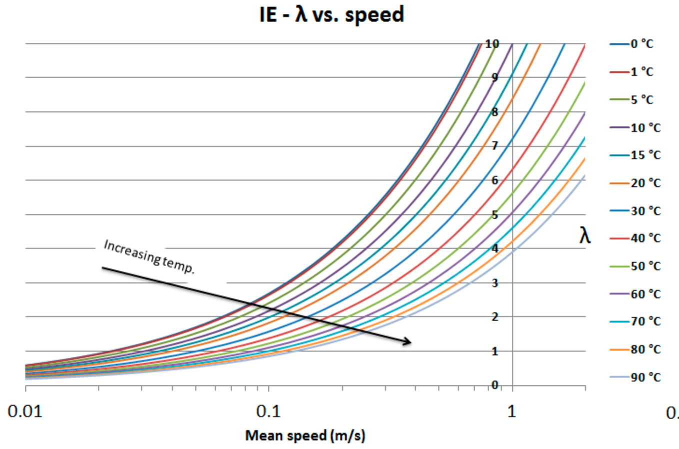
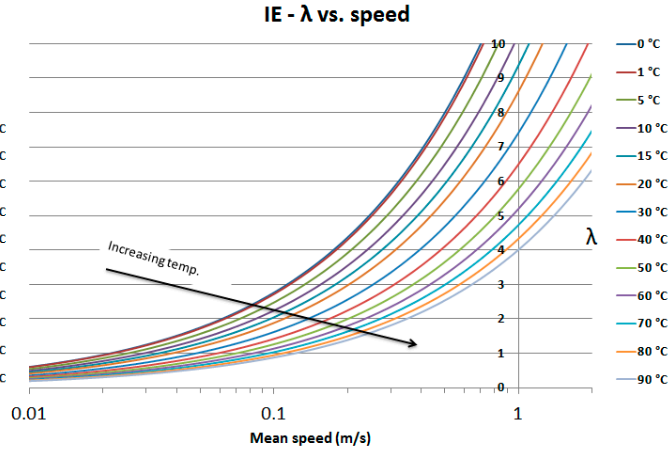


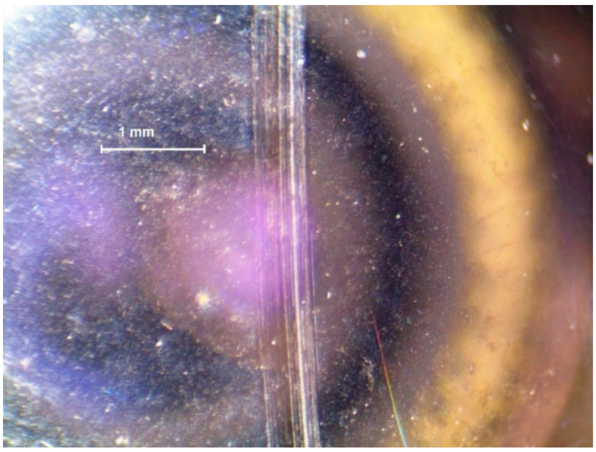
Figure SD2-B. Left plot: Calculated Stribeck parameter (λ) *vs.* mean speed compensated for the difference in Young's modulus caused by the change in temperature. Right plot: Calculated Stribeck parameter (λ) *vs.* mean speed not compensated for the difference in Young's modulus caused by the change in temperature.



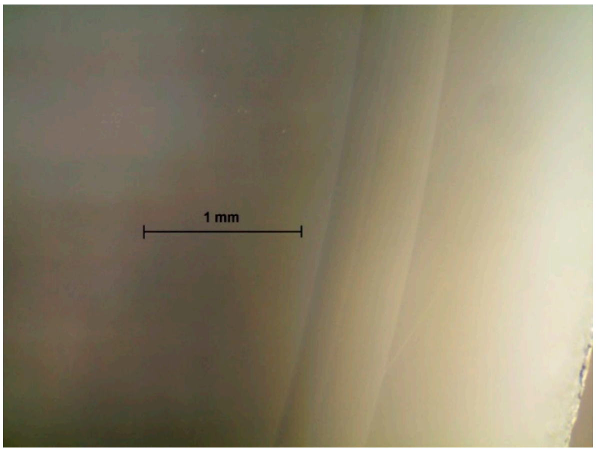


SD 3. Typical Wear Tracks of Hard Tribopairs

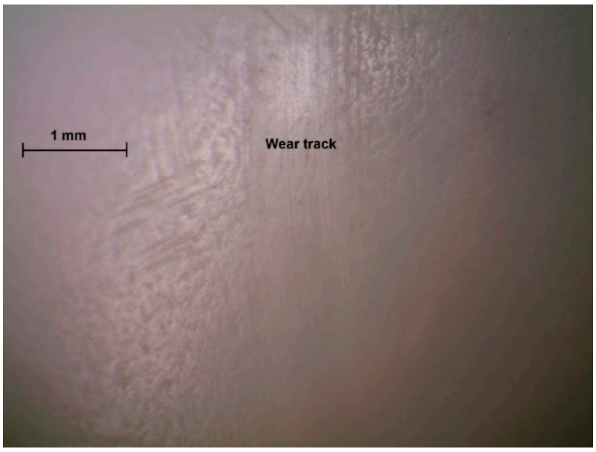
SD3-1. MTM 52100 Steel Ball.



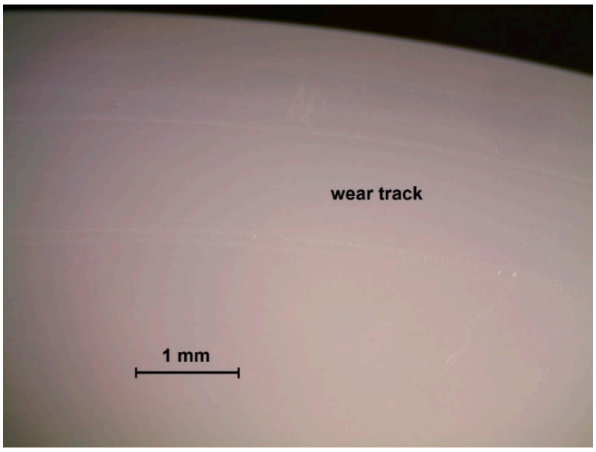
SD3-2. MTM Glass Disc.



SD3-3. MTM POM ball.



SD3-4. MTM POM disc.



SD 4. Theoretical Calculation of μ in Full-Fluid Film Hydrodynamic Regime of PDMS *vs.* PDMS Contact

A way to estimate the friction coefficient in hydrodynamic regime is to consider that the friction is generated solely from liquid shear between two solid surfaces.

The shear strength (σ) when shearing a (Newtonian) liquid is:

$$\sigma = \frac{du}{dy} = \frac{\frac{dx}{dt}}{dy} \cdot \eta \qquad (8)$$

where dx/dt is equal to the mean speed in the x direction, and dy is equal to the film thickness.
The shear strength (σ) is also equal to (where A is the area of contact/shear):

$$\sigma = \frac{F_{shear}}{A} \tag{9}$$

Hence

$$F_{shear} = \sigma \cdot A \tag{10}$$

Substituting Equation (8) into (10) we get:

$$F_{shear} = \frac{\frac{dx}{dt}}{dy} \cdot \eta \cdot A \tag{11}$$

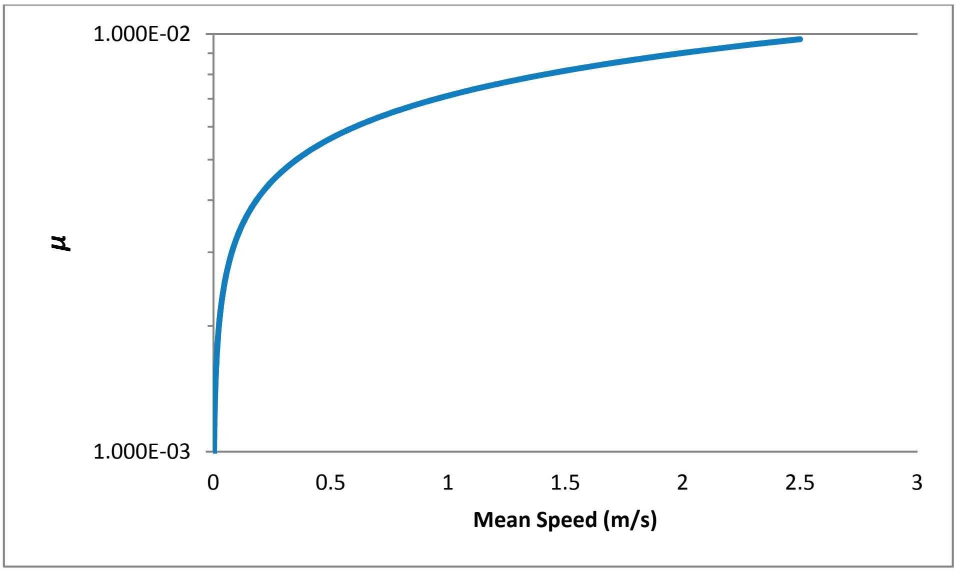
And then COF is:

$$\mu = \frac{F_{shear}}{F_{load}} = \frac{\frac{\frac{dx}{dt}}{dy} \cdot \eta \cdot A}{F_{Load}} \tag{12}$$

In Equation (12) we can insert the mean speed as dx/dt, and use the film thickness calculated by the Hamrock-Dowson equation for dy (dependent on speed). The viscosity of water is 1.75 mPas at 1 °C, and the area of contact for the PDMS-PDMS contact at 5 N load is 2.1×10^{-5} m^2. By using these parameters we can plot μ *vs.* mean speed for water at 1 °C:

As observed in Figure SD3, the friction coefficient is less than 0.008 even at 1.2 m/s (1200 mm/s), hence the friction increase due the hydrodynamic shearing of water would not be observed in the operating speed range of 10–1200 mm/s even at 1 °C where the viscosity of water is largest.

Figure SD3. Calculated μ from shearing of water layers between PDMS *vs.* PDMS surfaces at 1 °C.



References

1. Rubinstein, M.; Colby, R.H. *Polymer Physics*; Oxford University, Press, Inc.: Oxford, UK, 2003; p. 259.
2. Wong, E.J. Modeling and Control of Rapid Cure in Polydimethylsiloxane (PDMS) for Microfluidic Device Applications, Modeling and Control of Rapid Cure in PDMS for Microfluidic Device Applications. Ph.D. Thesis, MIT, Boston, MA, USA, 1 September 2010.